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SUMMARY

The effects of selected alloying elements on the high-temperature creep and tensile strength and the ductile-brittle transition behavior of arc- and electron-beam-melted tungsten were investigated. The tensile and creep strength of arc- and electron-beam-melted tungsten above 2500° F were significantly increased, in order of decreasing effectiveness, by additions of boron, hafnium, tantalum, columbium, and rhenium. Additions of 2.11 and 6.64 atomic percent rhenium to electron-beam-melted tungsten resulted in bend-transition temperatures below room temperature in the stress-relieved condition compared with 180° to 250° F for unalloyed arc- and electron-beam-melted tungsten.

INTRODUCTION

The need for structural materials with useful strength above 2500° F has stimulated interest in refractory-metal alloys. Because of its high melting point, tungsten appears to be one of the most attractive base metals for these alloys. One of the major deterrents to the use of tungsten materials has been their relatively high ductile-brittle transition temperatures, which are generally well above room temperature (typically 300° to 800° F). This study was made in an effort to identify tungsten-alloy systems that yield high strengths above 2500° F and, if possible, improved low-temperature ductility.

Previous investigations have identified some promising tungsten-alloy systems. References 1 and 2 showed that additions of tantalum and columbium were effective high-temperature strengtheners of tungsten; reference 3 shows that the addition of zirconium, vanadium, and carbon to a tungsten 12-weight-percent columbium alloy produced an alloy with a tensile strength at 3500° F, more than five times that of unalloyed tungsten. Research on powder-metallurgy tungsten alloys has identified dispersoid additions such as thorium oxide (ref. 4) as effective strengtheners. The strengthening effect of the thoria additions has been attributed largely to the retention of cold work to higher temperatures than possible with unalloyed tungsten (ref. 5).

Improvements in the low-temperature ductility of tungsten have been

reported by various investigators; the most striking have been shown by binary tungsten-rhenium alloys. Additions of rhenium of approximately 26 atomic percent produce room-temperature ductility (ref. 6), while dilute (3 to 7 atomic percent) tungsten-rhenium alloys have significantly lowered the ductile-brittle transition of tungsten (refs. 7 and 8). Dispersoid additions such as thoria and zirconia reduce the transition temperature of powder-metallurgy tungsten (ref. 8), while similar effects were observed with hafnium and carbon in electron-beam-melted tungsten sheet (ref. 9).

The purpose of this investigation was to study the effect of alloying on the high-temperature tensile and creep strength and low-temperature ductility of arc- and electron-beam-melted tungsten. Binary alloys in the tungsten-columbium, tungsten-tantalum, tungsten-rhenium, tungsten-hafnium, and tungsten-boron systems and ternary alloys of tungsten-columbium-carbon and tungsten-tantalum-boron were prepared and fabricated to rod and sheet to study these effects.

TABLE I. - ANALYSIS OF ARC- AND ELECTRON-BEAM-MELTED TUNGSTEN ALLOYS

Composition atomic percent	Interstitial content, ppm		
	Carbon	Nitrogen	Oxygen
Arc melted			
100W, AM-1	14	13	3
100W, AM-2	4	9	2
[a]100W, TM-1	4	--	5
W-0.83 Cb	2	--	3
W-0.99 Cb	5	--	8
W-2.22 Cb	--	--	--
W-3.22 Cb	4	--	5
W-1.25 Cb-0.54 C	350	--	--
W-1.15 Cb-1.32 C	850	--	--
W-0.88 Ta	12	18	100
W-2.46 Ta	3	17	38
W-3.53 Ta	3	18	14
W-5.44 Ta	6	--	--
W-1.00 Ta-0.22 B	5	--	3
W-3.0 Re	2	9	2
W-5.0 Re	7	46	3
W-0.44 Hf	5	25	21
W-0.25 B	4	5	4
W-0.59 B	5	8	8
W-0.67 B	9	10	4
W-1.69 B	--	--	--
Electron-beam melted			
100W, EB-1	5	24	2
100W, EB-2	8	5 - 13	3
100W, EB-3	15	27	2
W-2.11 Re	5	--	2
W-6.64 Re	8	--	5

[a]Melted near top of mold.

EXPERIMENTAL PROCEDURE

Melting

Tungsten-base alloys were prepared by vacuum-consumable-arc or electron-beam melting of pressed and sintered electrodes of tungsten and elemental alloying addition powders. Table I lists the alloys investigated in this program, with their interstitial analyses (obtained on the fabricated materials). Alloying additions were selected on the basis of high melting point and expected solid solution and/or dispersion-strengthening behavior. The arc-melted alloys contained up to 3.22 atomic percent columbium, 5.44 atomic percent tantalum, 0.44 atomic percent hafnium, 5 atomic percent rhenium, and 1.69 atomic percent boron, while the electron-beam-melted alloys contained up to 6.64 atomic percent rhenium. Limited studies were also made on arc-melted ternary tungsten-columbium-carbon and tungsten-tantalum-boron alloys. Arc melting was performed by using direct-current straight polarity (electrode negative) into a 2- or 2.68-inch-diameter water-cooled copper crucible. The arc-melting

facility is described in reference 1. One unalloyed tungsten ingot (TM-1, table I) was melted by using a mold with a retractable stool. With this technique, melting took place near the top of the mold resulting in a greater exposure of the molten pool to the vacuum. It was expected that this would result in higher purity due to the more efficient removal of volatile impurities. All of the other arc-melted materials were melted with the conventional deep mold.

The electron-beam-melted alloys were prepared from similar electrode material. An initial rapid melt was used to consolidate the electrode into an ingot followed by one to five remelts into a 2.5-inch-diameter copper crucible with a retractable stool. The electron-beam-melting facility is described in reference 10. The chamber pressure observed during arc- and electron-beam melting was less than 10^{-5} millimeter of mercury. The slower melt rates and greater exposure to the vacuum in electron-beam melting resulted in purer materials than those obtained by arc melting, as will be pointed out in subsequent discussion.

Fabrication

Table II lists the fabrication conditions for all the materials in this study. Both the arc- and electron-beam-melted ingots were first broken down by extrusion at temperatures of 2700° to 4800° F and reduction ratios of 6 or 8. Both conventional hydropress and high-energy Dynapak extrusion techniques were employed, as shown in table II. Approximately 14 percent of the alloys were extruded by using sintered molybdenum extrusion cans, while the remaining alloys were extruded in the unclad condition. It was expected that the molybdenum can would result in greater lubricity and subsequently greater ease of extrusion. Although no quantitative data are available yet, it has been observed that a greater percentage of the high-strength tungsten alloys were capable of being extruded by this technique. The extrusions were induction preheated in hydrogen by using a tungsten susceptor and were worked an average of 15 percent per pass in both swaging and rolling. Most of the arc- and electron-beam-melted alloys were fabricable by these techniques, although alloys containing approximately 1 and 2 atomic percent zirconium (not listed in table II) were successfully extruded but could not be rolled or swaged.

Mechanical Testing

Swaged rod was ground into buttonhead tensile specimens having a gage length of 1.03 inch and a 0.16- or 0.14-inch reduced diameter, while longitudinal specimens measuring 0.3 by 0.9 inch were cut from 0.05-inch-thick sheet for bend testing. For tests below 1000° F, the tensile and bend specimens were electropolished in a 2-percent sodium hydroxide solution to remove at least 5 mils from the surface. Tensile testing was conducted in vacuum in a tensile testing machine at a crosshead speed of 0.005 inch per minute to yield, and 0.05 inch per minute to fracture. Heating below 1000° F was conducted in a platinum-wound resistance furnace, while heating at 2500° F and higher was accomplished with a tantalum-sleeve resistance heater in a water-cooled

TABLE II. - FABRICATION CONDITIONS FOR ARC-MELTED AND ELECTRON-BEAM-MELTED TUNGSTEN ALLOYS

Alloy composition, atomic percent	Extrusion temperature, °F	Reduction ratio	Extrusion constant, K,[a] lb/sq in.	Average swaging and rolling temperature, °F	Percent reduction in swaged rod	Percent reduction in rolled sheet
Arc melted						
100W, AM-1	3500	6	93,600	2200 - 2650	66.8	(b)
100W, AM-2	3450	6	92,400	2500	82.6	---
100W, TM-1	3600	8	[c]65,300	2200	69.7	(b)
W-0.83 Cb	3600	6	67,200	2550	74.4	----
W-0.99 Cb	3500	6	78,400	2600	83.0	92.3
W-2.22 Cb	3700	8	Dynapak	----	----	----
W-3.22 Cb	3800	6	Instrumentation failed	2500 - 2600	79.6	(b)
W-1.25 Cb-0.54 C	4800	6	81,200	----	----	---
W-1.15 Cb-1.32 C	4500	6	103,000	----	----	---
W-0.88 Ta	3500	6	96,600	2600	68.3	88.6
W-2.46 Ta	3550	8	Dynapak	2700	66.6	(b)
W-3.53 Ta	3800	6	Instrumentation failed	2700	79.6	96.3
W-5.44 Ta	3800	6	94,100	----	----	----
W-1.0 Ta-0.22 B	2700	8	Dynapak	2500	66.6	----
W-3.00 Re	4000	8	[c]62,400	2600	76.8	(b)
W-5.00 Re	4000	8	[c]86,400	2700	76.8	(b)
W-0.44 Hf	3800	8	Dynapak	2650	82.6	(b)
W-0.25 B	3500	8	Dynapak	2500	77.6	(b)
W-0.59 B	3800	8	Dynapak	2700	77.6	---
W-0.67 B	4000	6	[c]67,600	2600	80.0	(b)
W-1.69 B	4180	6	Partial sticker	2650	----	(b)
Electron-beam melted						
100W, EB-1	3000	6	90,400	2100	86.9	(b)
100W, EB-2	3500	6	70,400	2200	46.0	---
100W, EB-3	3000	6	(c)	2400	83.0	(b)
W-2.11 Re	3400	6	[c]67,800	2450	86.9	(b)
W-6.64 Re	3400	8	[c]79,800	2400	83.0	(b)

[a]Extrusion constant K calculated from formula: $K = p/\ln R$, where p is breakthrough pressure and R is area reduction ratio.

[b]Rolled from 0.250 in. to 0.050 in. after intermediate stress-relief anneal. Total reduction from extrusion was approximately 92 percent.

[c]Clad with molybdenum.

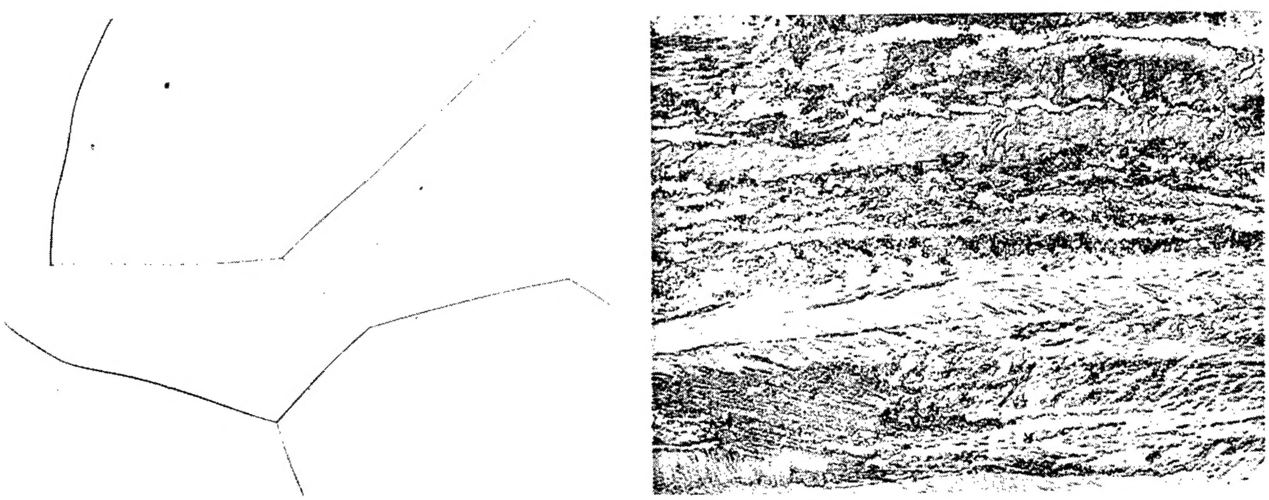
stainless-steel shell. Temperature was recorded with a platinum - platinum-13-percent-rhodium thermocouple below 1000° F and with a tungsten - tungsten-26-percent-rhenium thermocouple above 2500° F. Further details of the testing procedure are given in reference 10. Creep tests were conducted in a conventional beam-loaded machine with a heating setup similar to that for the short-time tensile tests. Specimen strain was assumed to be equal to the loading rod movement.

Bend testing of sheet electropolished to a thickness of approximately 0.040 inch was conducted at 2 inches per minute by using a bend radius of 4t (where t is the sheet thickness). The bend fixture is described in reference 11. Heating was performed in air in a platinum-wound resistance furnace, the temperature being recorded with a Chromel-Alumel thermocouple attached to the fixture near the specimen. Temperature measurements were believed accurate to ±5° F. The bend ductile-brittle transition temperature was defined as the median between the temperatures at which a complete bend and a brittle failure was obtained.

RESULTS AND DISCUSSION

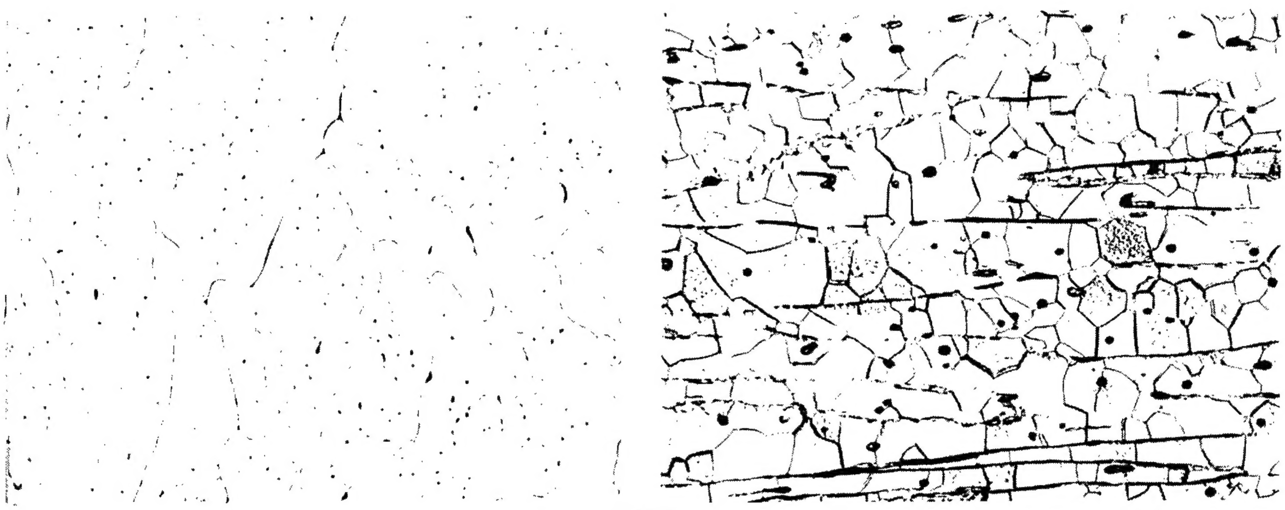
Melting and Fabrication

Figures 1 and 2 show microstructures of as-melted and as-extruded single- and two-phase alloys. The electron-beam-melted alloys exhibited a partially recrystallized structure after extrusion, while the arc-melted alloys showed a variety of microstructures ranging from fully recrystallized to fully worked. The extrusion microstructure could not be correlated with any of the extrusion variables, including composition. All of the Dynapak extrusions, however, showed full recrystallization regardless of prior melting process.



(a) As melted. X250. C-71925 (b) As extruded. X150.

Figure 1. - Microstructures of arc-melted tungsten 3.53 atomic percent tantalum. Electropolished; etched with boiling 3 percent hydrogen peroxide.



(a) As melted. X100. C-71926 (b) As extruded; note oriented borides. X150.

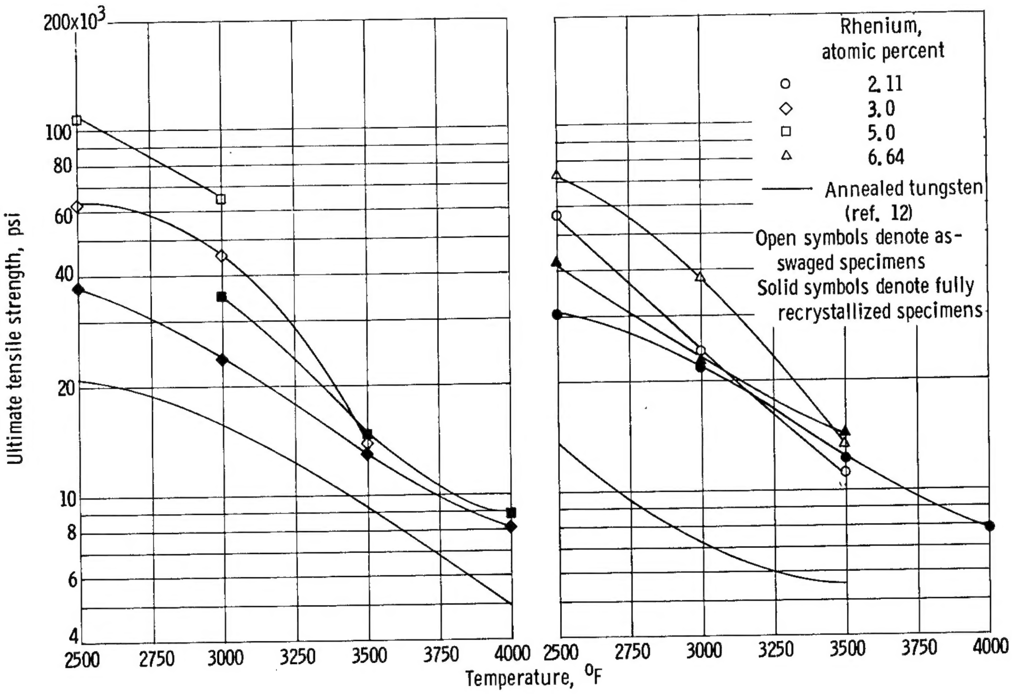
Figure 2. - Microstructures of arc-melted tungsten 0.67 atomic percent boron. Electropolished; etched with boiling 3 percent hydrogen peroxide.

Figure 2(a) is typical of the boride- or carbide-containing alloys. The second phase formed either heavy grain-boundary networks or discrete intragranular particles. After extrusion, the borides and carbides were oriented in the extrusion direction, as shown in figure 2, implying that no dissolution of the borides took place during the extrusion.

High-Temperature Tensile Properties

The effect of temperature on the ultimate tensile strength of selected binary tungsten-alloy systems is plotted semilogarithmically in figures 3 to 5. Included are alloys in the tungsten-columbium, tungsten-tantalum, tungsten-rhenium, tungsten-hafnium, and tungsten-boron binary systems. Data are presented for alloys in the extruded or swaged condition and after a 1-hour anneal at 3200° or 3600° F. The unalloyed tungsten data are average values for annealed materials from reference 12.

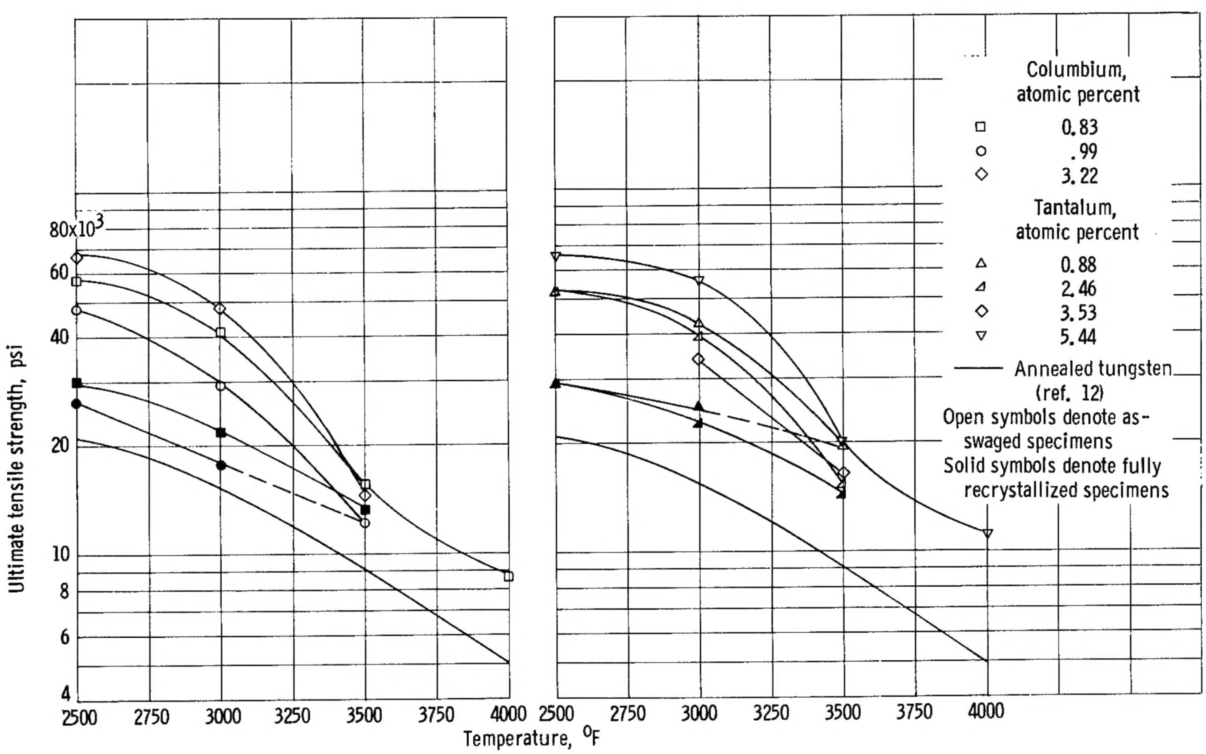
The strengthening effect of prior warm work is shown in figure 3 by a comparison of the properties of arc- and electron-beam-melted tungsten-rhenium alloys. Prior warm work was an effective strengthener at 2500° F but at higher temperatures the effect varied. Arc-melted alloys showed a strengthening effect at 3000° F, but at 3500° F, little difference remained between the annealed and worked materials. Electron-beam-melted alloys, on the other hand, showed less effect of warm work at lower temperatures. One of the two electron-beam-melted alloys tested (2.11 atomic percent Re) showed no warm work strengthening at 3000° F. This behavior is similar to that reported previously for unalloyed arc- and electron-beam-melted tungsten (ref. 12). The loss of the warm-work strengthening at high temperatures was due to recrystallization during the test. The electron-beam-melted alloys, because of their higher purity, recrystallized in a lower temperature range (ref. 12) and therefore do not retain their worked structures to as high a temperature as the arc-melted alloys.



(a) Arc melted.

(b) Electron-beam melted.

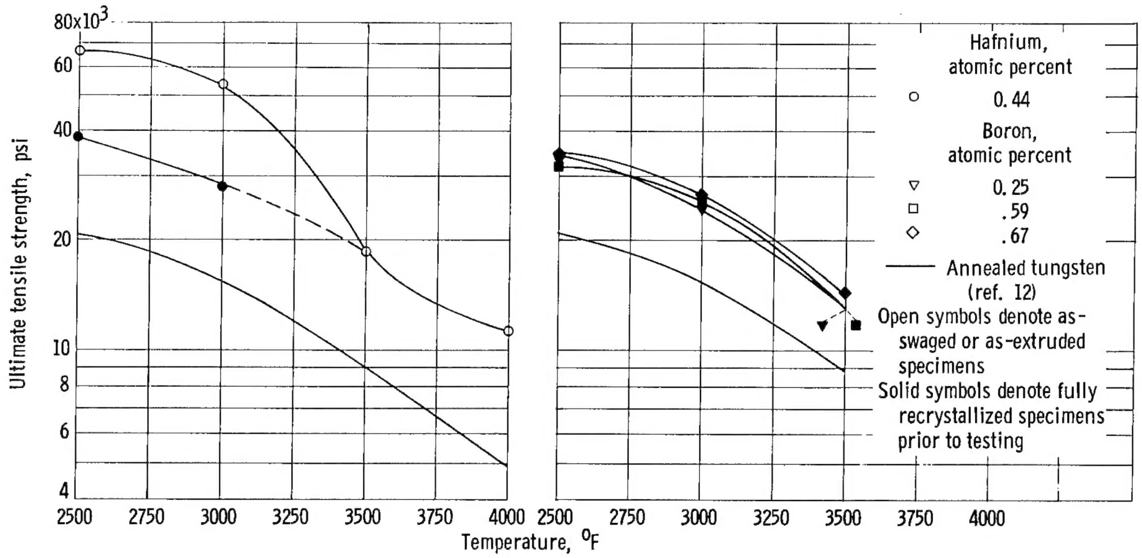
Figure 3. - Effect of temperature on ultimate tensile strength of arc- and electron-beam-melted tungsten-rhenium alloys.



(a) Tungsten-columbium.

(b) Tungsten-tantalum.

Figure 4. - Effect of temperature on ultimate tensile strength of arc-melted tungsten-columbium and tungsten-tantalum alloys.



(a) Tungsten-hafnium.

(b) Tungsten-boron.

Figure 5. - Effect of temperature on ultimate tensile strength of arc-melted tungsten-hafnium and tungsten-boron alloys.

Figures 4 and 5 show the effect of temperature on the strength of arc-melted tungsten-tantalum, tungsten-columbium, tungsten-hafnium and tungsten-boron alloys. The general comments concerning retention of warm work discussed previously apply here also. A boron addition (fig. 5) of 0.25 atomic

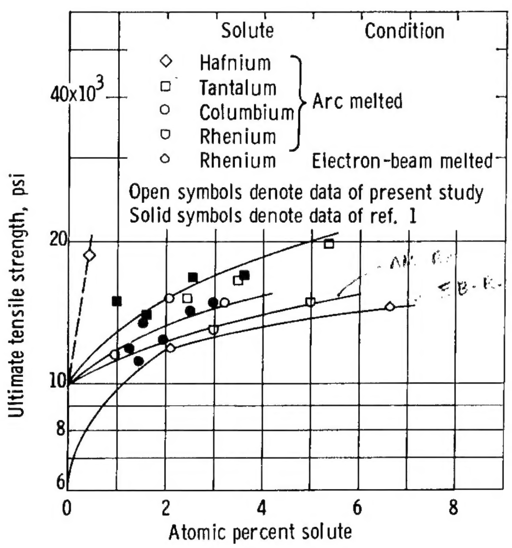


Figure 6. - Effect of alloying on 3500° F ultimate tensile strength of arc- and electron-beam-melted tungsten.

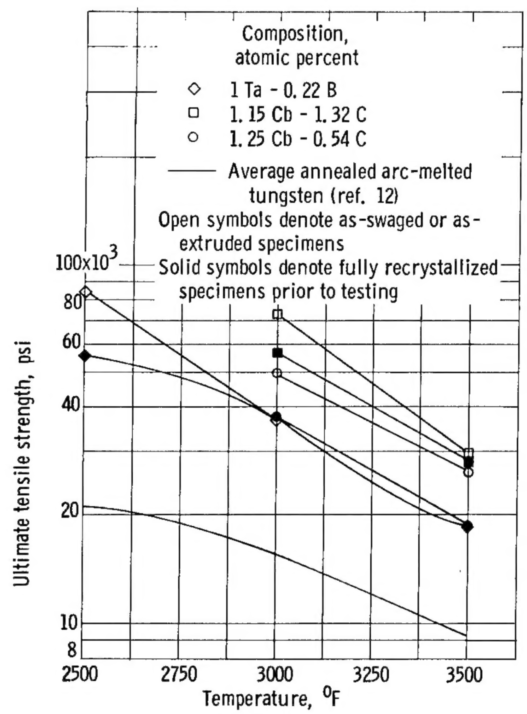


Figure 7. - Effect of temperature on ultimate tensile strength of ternary tungsten-columbium-carbon and tungsten-tantalum-boron alloys.

percent produced a rapid increase in the tensile strength in the annealed condition, but larger additions produced only moderate additional strengthening.

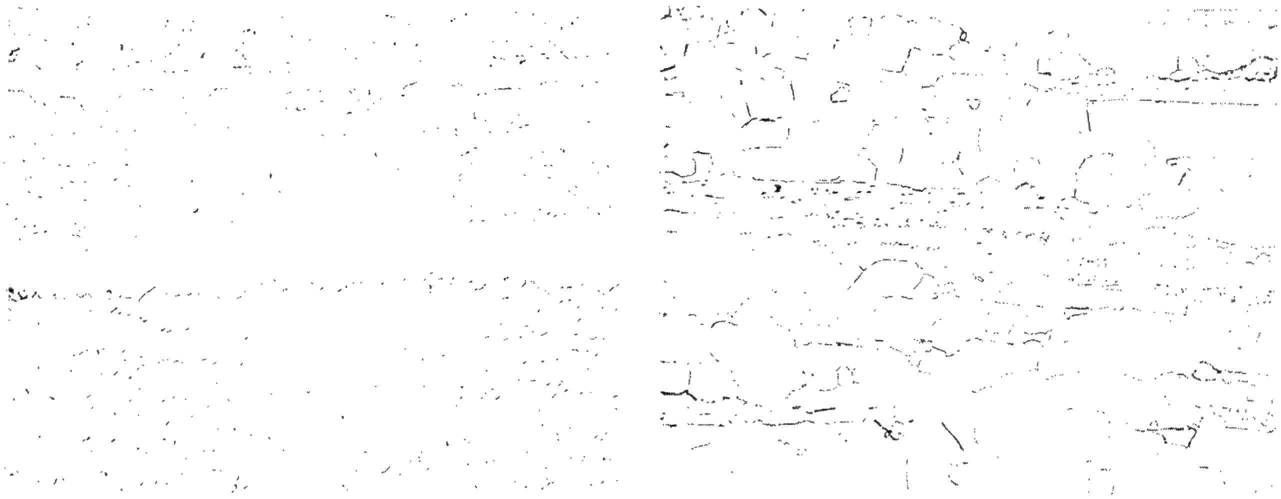
Figure 6 summarizes the effect of the binary alloy additions on the ultimate tensile strength at 3500° F for arc- and electron-beam-melted alloys. Additional data for this plot were taken from reference 1. The scatter in the tensile data for the tungsten-columbium and tungsten-tantalum alloys at low alloying levels was attributed primarily to variation in their interstitial contents. The effect of small changes in interstitial content on strength is illustrated by the tungsten 0.88-atomic-percent-tantalum alloy (not shown in fig. 6) whose tensile strength is shown in figure 4. This alloy had a strength of 19,420 psi at 3500° F, compared with a strength of approximately 13,000 psi expected with this tantalum content by interpolation of the plot of strength against tantalum content in figure 6. Subsequent chemical analysis of the alloy revealed that it contained an abnormally high oxygen content of 100 ppm (0.11 atomic percent). The additional strengthening in this alloy is attributed to a fine dispersion of tantalum oxide.

Data for the other binary alloys in figure 6 show that the order of effectiveness at 3500° F of alloying elements in improving the strength of arc-melted tungsten is hafnium, tantalum, columbium, and least effective, rhenium. This order also holds for annealed materials at 2500° and 3000° F.

Ternary tungsten-columbium-carbon and tungsten-tantalum-boron alloys (fig. 7) had strengths superior to the binary columbium and tantalum alloys. For example, a tungsten 1.15-atomic-percent-columbium 1.32-atomic-percent-carbon alloy in the annealed condition had a strength at 3500° F that was more than double that of the nominal tungsten 1-atomic-percent-columbium alloys. The tungsten 1-atomic-percent-tantalum 0.22-atomic-percent-boron alloy was nearly equivalent in strength to the tungsten 1.25-atomic-percent-columbium 0.54-atomic-percent-carbon alloy with an ultimate tensile strength of 18,600 psi at 3500° F. At 3000° F, the extruded tungsten 1.15-atomic-percent-columbium 1.32-atomic-percent-carbon alloy had a tensile strength of 72,600 psi. This is believed to be the highest strength yet reported for a metallic material at this temperature.

(W-1.25 % Cb-.54 % C ; W-1.15 % Cb-1.32 % C)

Subsequent light and electron microscopy revealed that the two carbon-containing alloys were only partially recrystallized during extrusion at 4500° to 4800° F and contained a dispersion, as shown in figure 8. X-ray analysis revealed that the dispersion was tungsten carbide (W_2C) although a slight shift in the lines suggested that columbium was also dissolved in the carbide. The carbides were distributed within the grains and as a relatively coarse network at the grain boundaries. Electron microscopy revealed intragranular carbides as fine as 0.5 micron. After testing at 3500° F, the major structural change appeared to be the formation of a substructure within the grains in the reduced section, an example of which is shown in figure 9. The appearance after testing suggests that a substructure was formed during testing and stabilized by the fine carbide dispersion, thus contributing substantially to the high strengths observed. These data reveal that the high strength of these alloys is derived primarily from a stabilization of the cold-worked structure by the carbide particles.



(a) Tungsten 1.25-atomic-percent-columbium 0.5-atomic-percent-carbon alloy.

C-71927

(b) Tungsten 1.15-atomic-percent-columbium 1.32-atomic-percent-carbon alloy.

Figure 8. - Extrusion microstructures of tungsten-columbium-carbon alloys. Lactic-nitric-hydrofloric acid etchant. X250.

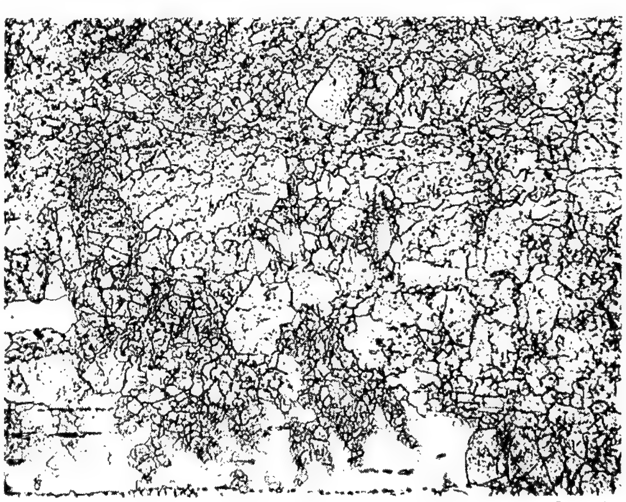
High-Temperature Creep Properties

Step-load creep data at 3500° F have also been obtained on selected alloys and typical data are shown in figure 10. The stress and linear creep rate were found to obey the relation

$$\dot{\epsilon} = K\sigma^n$$

where $\dot{\epsilon}$ is the linear creep rate, σ the stress, and K and n are constants. The stress dependence of the linear creep rate n varied from 5.3 to 6.1 compared with the value of 5.8 for unalloyed tungsten (ref. 12).

Figure 11 is a plot of the stress at a creep rate of 10^{-6} per second, corresponding to a rupture life of approximately 50 hours at 3500° F, as a function of solute content. The order of effectiveness of the various alloying additions is the same as that for the short-time tensile properties, suggesting that the strengthening mechanisms are the same or similar for both creep and short-time tensile behavior of the alloys in this investigation.



C-71928

Figure 9. - Microstructure of tungsten 1.25-atomic-percent-columbium 0.54-atomic-percent-carbon alloy after annealing at 3600° F and testing at 3500° F. Sodium hydroxide electrolytic etchant. X250.

The observed effectiveness of these alloying additions on the

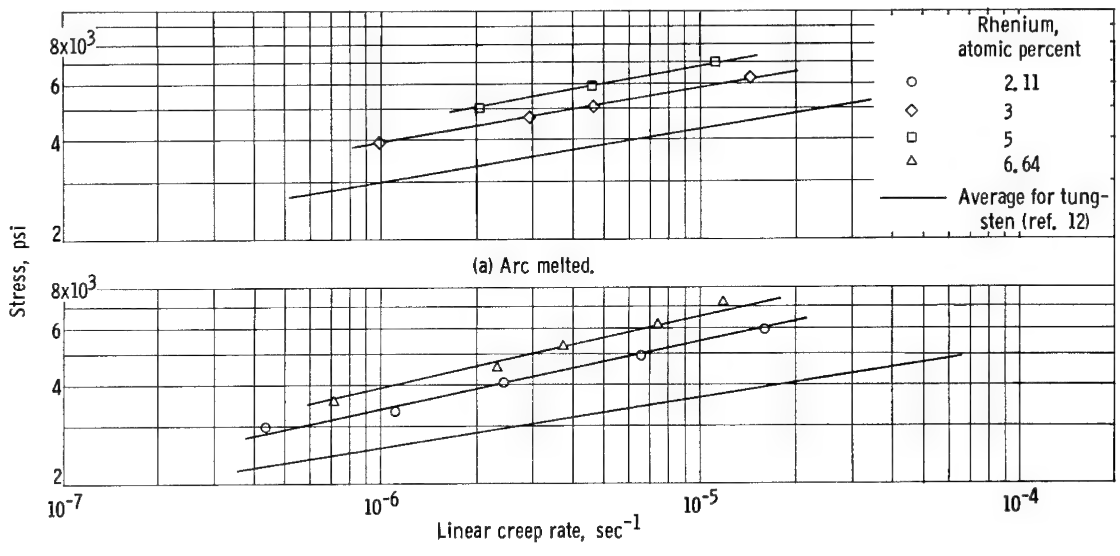


Figure 10. - Stress dependence of linear creep rate for arc- and electron-beam-melted tungsten-rhenium alloys at 3500° F in recrystallized condition.

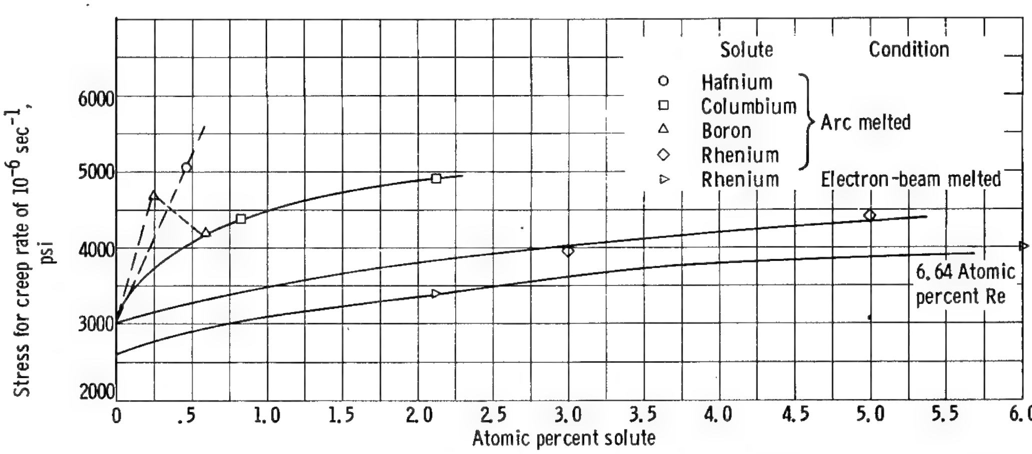


Figure 11. - Effect of alloying on creep strength of arc- and electron-beam-melted tungsten alloys at 3500° F.

high-temperature tensile and creep strength of tungsten can be rationalized in terms of the effects of alloying on the strain-hardening and recovery processes. The ultimate tensile strength represents the strengthening due to strain-hardening combined with the weakening by recovery that occurs during the duration of the tensile test, while second-stage creep represents a balance between strain-hardening and recovery processes during creep.

Examination of the present results in terms of dislocation concepts assists in understanding the role of alloying on strain-hardening and recovery processes, although no direct observations of the dislocation structure of these materials were made. The relative weakness of the electron-beam-melted alloys compared with the arc-melted alloys can be explained, in part, by an increase in the overall rate of thermal recovery and the coarser recrystallized

grain sizes as a result of the increased purification by electron-beam melting. Impurities have been shown to reduce the rate of recovery of metals even at temperatures where dislocation climb is rate-controlling. This could result from impurity atoms being bound to dislocation jogs, thus blocking their movement (ref. 13). Additional evidence is seen in electron-beam-melted tungsten where the recrystallization temperature has been shown to be 500° to 600° F lower than that in arc-melted tungsten (unpublished NASA data obtained by W. R. Witzke of Lewis). This can be taken as evidence for an increase in the rate of recovery. The coarser grain sizes of the electron-beam-melted materials probably have the effect of decreasing the rate of work-hardening (ref. 12).

The alloy additions investigated in this study presumably strengthened in a twofold manner, that is, by acting as solid-solution strengtheners and by reacting with residual interstitials to form fine dispersoids. Differences in atomic size between the base-metal and solute atoms have been previously shown to correlate the strengthening effect of solid-solution additions (ref. 14). The atomic sizes of tungsten and the alloy additions investigated in this study (ref. 15) are compared in the following table:

Element	Atomic radii, angstroms	Atomic size difference
Tungsten	1.38	0.00
Rhenium	1.34	.04
Tantalum	1.43	−.05
Columbium	1.44	−.06
Hafnium	1.55	−.17

Goldschmidt At. Radius	Size diff rel to W
1.41	0.0
1.38	.03
1.47	.06
1.47	.06
1.59	.18
Mo – 1.40	.01

A rough correlation is seen between the difference in atom size and the order of effectiveness in promoting high-temperature strength mentioned earlier. Differences in atom size would be expected to hamper climb by attachment of solute atoms to dislocation jogs and thus to decrease the rate of thermal recovery (ref. 12).

The role of tantalum, columbium, and hafnium in forming fine dispersoids is an additional strengthening factor. The results for the tungsten-columbium-carbon alloys suggest that the precipitates had a large effect on stabilizing the dislocation substructure at high temperatures, as did oxygen in the tungsten-tantalum alloys. The weak dispersion-forming tendency of rhenium, its inability to form a carbide (ref. 16), and its small atomic size difference reduce its strengthening effect in tungsten at elevated temperatures.

Figure 12 is a summary of the tensile properties of the strongest tungsten-base alloys produced. The tungsten-columbium-zirconium-vanadium-carbon alloys at 3500° F (ref. 3) and the tungsten 1.15-atomic-percent-columbium 1.32-atomic-percent-carbon alloy of this study are the strongest alloys yet reported in the literature. It is of interest to note that the strongest alloys for use at 3500° F and above, such as the tungsten-thoria, tungsten-columbium-zirconium-vanadium-carbon, and tungsten-columbium-carbon alloys, are those in which dispersion-strengthening plays a predominant role. It is known, however, that dispersion-strengthening in these systems is strongly influenced by processing conditions, such as fabrication temperature and

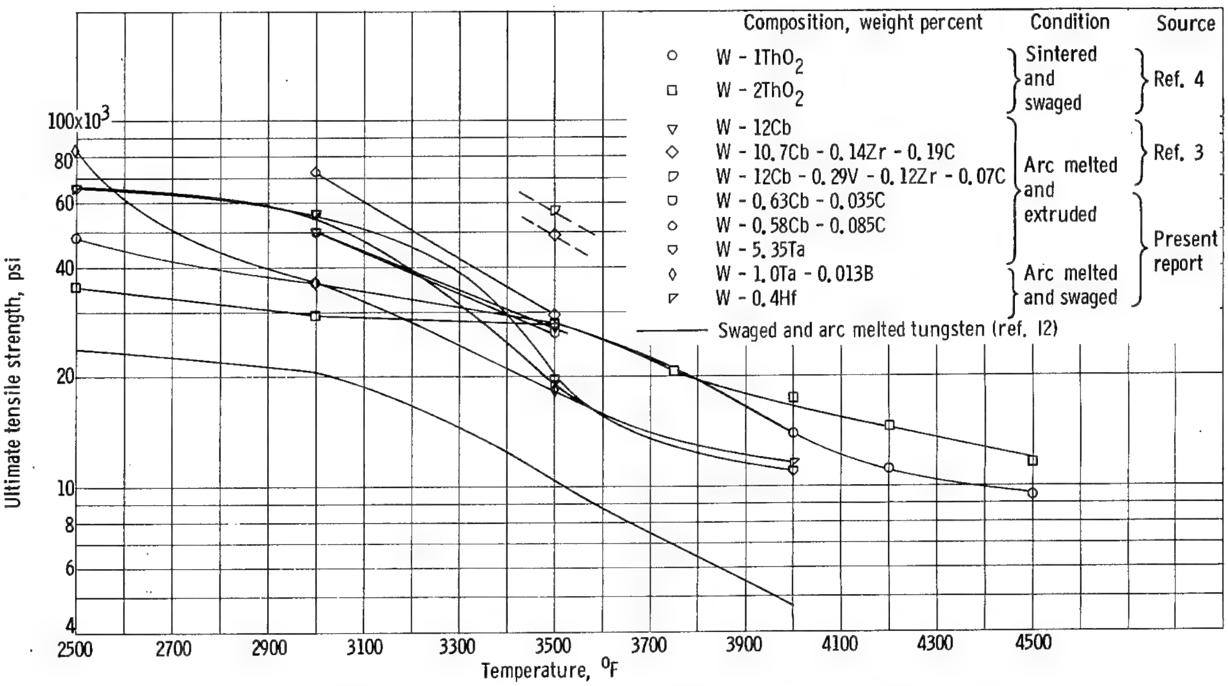


Figure 12. - Effect of temperature on strength of various tungsten-base alloys.

degree of deformation, which may affect the size and distribution of the dispersed phase. Optimization of process variables to maximize dispersion-strengthening will lead to stronger tungsten-base alloys in the future.

Low-Temperature Tensile and Bend Properties

The ductile-brittle transition behavior of arc- and electron-beam-melted tungsten and tungsten alloys was studied both in tension and bending. Tensile tests were conducted on electropolished rod in the recrystallized condition, while bend tests were made with electropolished sheet (0.040 in. thick) in the as-rolled, stress-relieved, and fully recrystallized conditions.

Figure 13 shows the effect of temperature on the tensile ductility of recrystallized arc- and electron-beam-melted tungsten after various annealing treatments. The three lots of arc-melted tungsten exhibited similar transition behaviors and the data could be represented by a narrow band, as shown in the figure. On the other hand, the transition behavior of the electron-beam-melted tungsten varied greatly with annealing temperature. Of particular interest is that the transition temperature (arbitrarily defined for the tensile test as the temperature at 40 percent reduction in area) of electron-beam-melted tungsten decreased with increasing annealing temperature in the range 2500° to 4000° F. This anamolous behavior is believed to be a result of the rapid grain growth above the recrystallization temperature that results in large-grained structures containing only 3 to 17 grains in the specimen cross section. Although the reasons for this behavior are not yet clear, similar results have been observed in chromium (ref. 17). The lower transition temperature of very

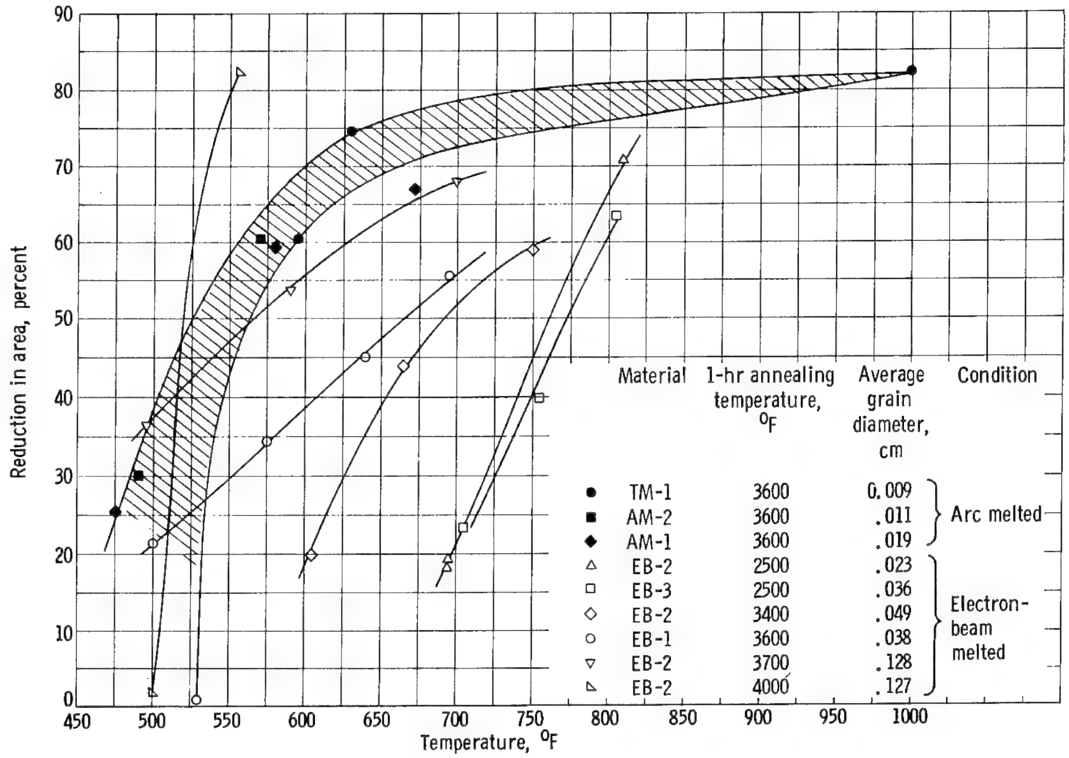


Figure 13. - Effect of temperature on ductility of arc- and electron-beam-melted tungsten.

coarse-grained material was attributed to a decrease in the probability of grain-boundary crack initiation by a more favorable distribution of stresses within the grains due to the large grain sizes. Other studies have shown that for powder-metallurgy (ref. 18) and electron-beam-melted (ref. 19) tungsten with grain sizes less than about 0.01 centimeter, the normal decrease in the transition temperature with decreasing grain size does occur.

No correlation of ductility with purity between the arc- and electron-beam-melted materials was found in the present investigation. It is worthy of note, however, that although the annealing of electron-beam-melted tungsten at 3700° and 4000° F produced identical grain sizes of 0.128 centimeter, the ductility of the specimens annealed at 4000° F decreased much more sharply with decreasing temperature. This behavior may be due to differences in the impurity distributions at the grain boundaries after the different annealing treatments.

The effects of various alloy additions on the tensile transition behavior of recrystallized (1 hr at 3600° F) arc- and electron-beam-melted tungsten are shown in figure 14. Boron and hafnium additions were the most embrittling while rhenium had the least effect. In comparison, the effect of carbon and oxygen on the transition temperature determined on commercial tungsten rod (ref. 20) was more severe than the boron additions in the present investigation. For example, an addition of 60 ppm carbon raised the transition temperature from 450° to 780° F.

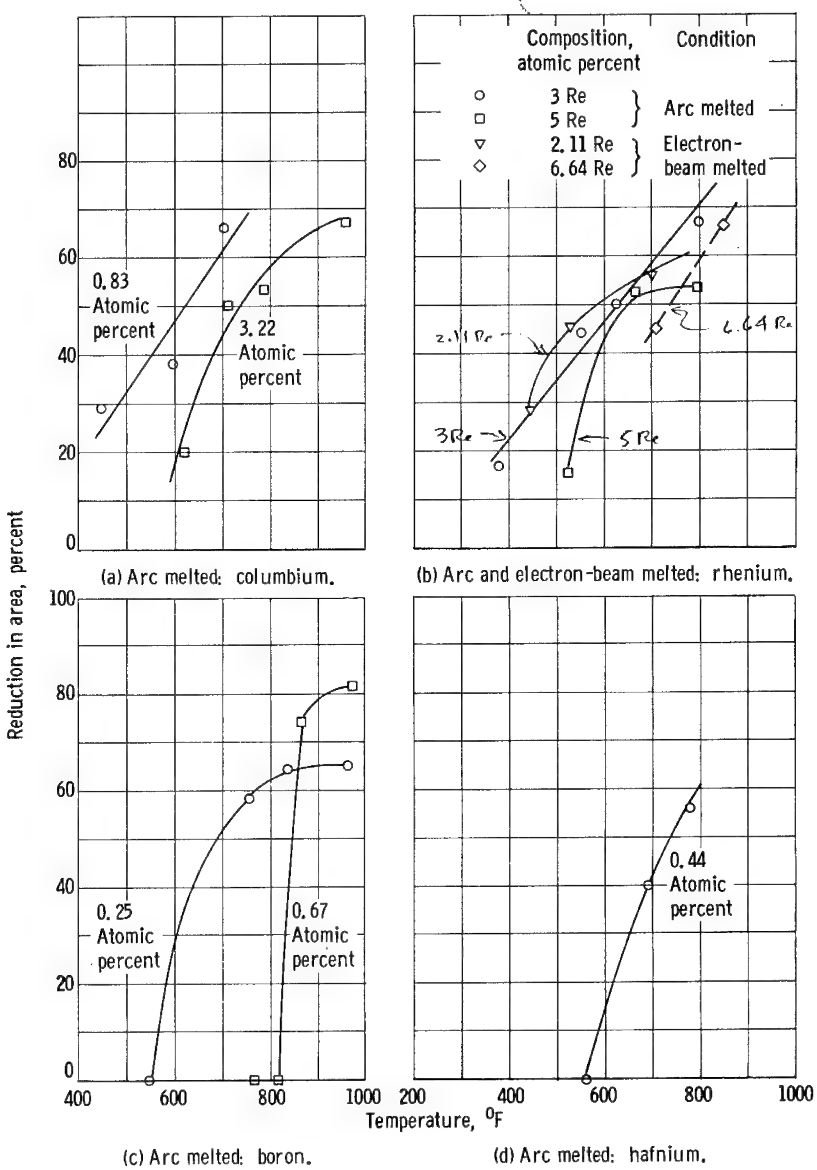


Figure 14. - Effect of temperature and alloying on ductility of recrystallized arc- and electron-beam-melted tungsten.

Bend transition tests (4t) were performed on 0.040-inch-thick electropolished strip rolled from the preceding and additional alloys. Data are given in table III for material in the as-rolled, stress-relieved, and fully recrystallized conditions for several alloys. For most alloys, only a small amount of sheet was available, and bend tests were conducted as fully recrystallized materials only. Transition temperatures for the as-rolled materials varied from 310° F for an arc-melted tungsten 0.99-atomic-percent-columbium alloy to less than room temperature (75° F) for an electron-beam-melted tungsten 6.64-atomic-percent-rhenium alloy. The effects of stress-relief annealing varied among the different alloys. For example, the previously mentioned transition temperature for the tungsten 0.99-atomic-percent-columbium alloy was lowered from 310° to 235° F after annealing at 2200° F for 1 hour, while annealing of a tungsten 3.53-atomic-percent-tantalum alloy under the same conditions raised the transition temperature from 212° to 262° F. After stress-relief annealing, the electron-beam-melted tungsten 2.11-atomic-percent-rhenium and tungsten 6.64-atomic-percent-rhenium alloys had transition temperatures of less than 75° F.

The bend-transition temperature for unalloyed tungsten in the fully recrystallized condition varied from 485° for electron-beam-melted and 570° F for top-of-the-mold arc-melted tungsten to about 670° F for deep-mold arc-melted tungsten. Differences in interstitial content and grain size do not appear to account for these differences in transition temperature. Figure 15 is a plot of the recrystallized bend-transition temperature against alloy com-

TABLE III. - EFFECT OF ALLOYING ON 4t BEND-TRANSITION TEMPERATURE OF ARC- AND ELECTRON-BEAM-MELTED TUNGSTEN

[Crosshead speed, 2 in./min]

Composition, atomic percent	As-rolled	Stress relieved; 2200° or 2400° F anneal	Recrystallized; 3400° or 3600° F anneal
Arc-melted			
100W, AM-1	---	---	660
100W, AM-2	218	255	685
100W, TM-1	195	---	570
W-3.0 Re	280	---	400
W-5.0 Re	240	---	375
W-0.99 Cb	310	235	670
W-3.22 Cb	---	---	605
W-0.88 Ta	185	200	---
W-2.46 Ta	---	---	650
W-3.53 Ta	212	262	575
W-0.44 Hf	---	---	655
W-0.25 B	---	---	670
W-0.67 B	---	---	470
W-1.69 B	---	---	490
Electron-beam melted			
100W, EB-1	195	---	485
W-2.11 Re	≤75	≤75	525
W-6.64 Re	≤75	≤75	775

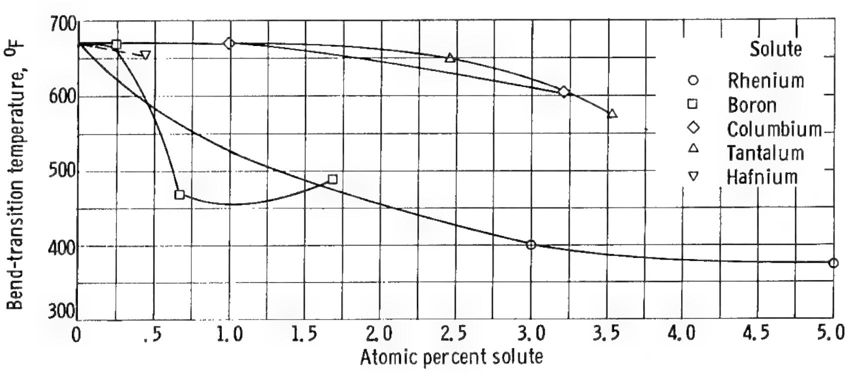


Figure 15. - Influence of alloying on bend-transition temperature of recrystallized arc-melted tungsten.

position. Additions of columbium, tantalum, and hafnium resulted in moderate decreases in the transition temperatures of recrystallized materials.

Significant decreases in the bend-transition temperature were observed for the recrystallized arc-melted tungsten-rhenium and tungsten-boron alloys, (fig. 15). The effect of rhenium was to lower the transition temperature from about 670° F for unalloyed tungsten to 400° F for an addition of 3-atomic-percent rhenium and to 375° F for the 5-atomic-percent-rhenium alloy. Similar decreases were not observed for the recrystallized electron-beam-melted tungsten-rhenium alloys (table III). Additions of boron of 0.67 and 1.69 atomic percent decreased the bend-transition temperature to approximately the same value of 470° to 490° F. This is in contrast to the tensile results which showed that boron significantly raised the ductile-brittle transition temperature.

The ductile-brittle transition temperature in body-centered-cubic metals is usually attributed to the steep temperature dependence of the yield and flow

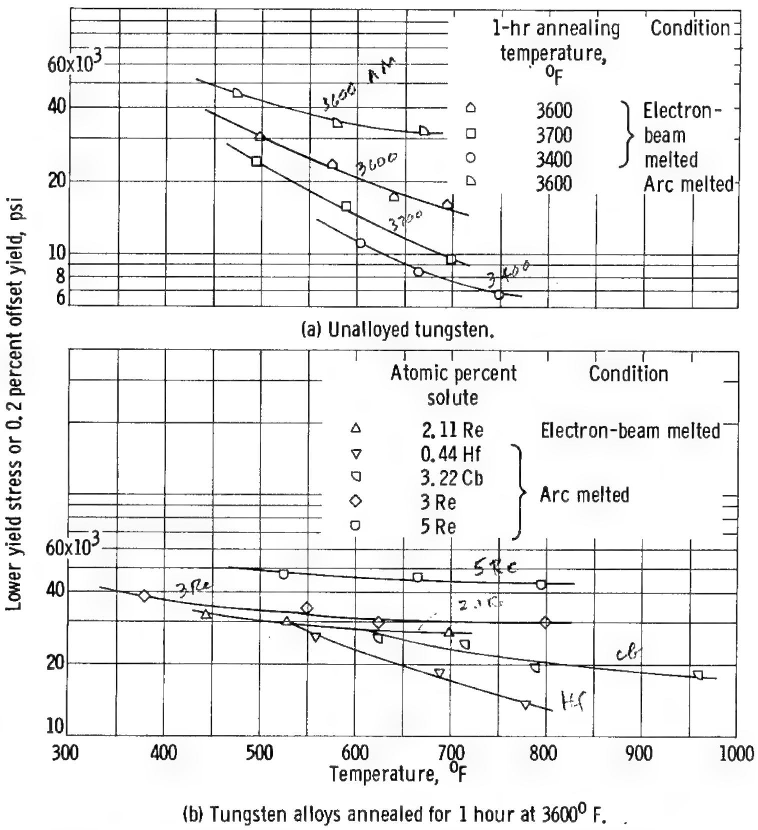


Figure 16. - Effect of temperature on yield strengths of arc- and electron-beam-melted tungsten and tungsten alloys.

stresses compared with the brittle-fracture stress (ref. 13). Low ductility values are observed at the temperature where the yield stress is comparable to the brittle-fracture stress. Figure 16 shows the tensile yield strengths for arc- and electron-beam-melted tungsten and selected alloys. The four lots of unalloyed tungsten all show a rapid increase in yield strength as the ductile-brittle transition temperature is approached as do the binary tungsten-columbium and tungsten-hafnium alloys. The tungsten-rhenium alloys show a significantly more gradual increase. The improved ductility observed in the dilute tungsten-rhenium alloys is considered to be associated with the decreased temperature dependence of the yield stress. Other investigators (ref. 21) have shown that the hardness of arc-melted dilute tungsten-rhenium alloys is lower than that of unalloyed tungsten at room temperature and below, but the situation is reversed at the higher temperatures. This can be taken as additional evidence of a decrease in the temperature dependency of the yield and flow stresses of dilute tungsten-rhenium alloys.

The observed decreases in the bend-transition temperature of annealed arc-melted tungsten by boron additions may be related to an increase in the volume fraction of the tungsten-boride particles, since all the additions were above the solubility limit of boron in tungsten (ref. 22). Additions of thoria and zirconia to powder-metallurgy tungsten also resulted in reduced transition temperature; this was attributed to a grain-refining effect (ref. 8). The tungsten-boron alloys studied in this investigation were considerably finer grained than unalloyed tungsten, but the grain size did not decrease for additions over 0.25 atomic percent boron, while the transition temperature decreased substantially. It is suggested that the presence of boride particles might impede the propagation of cracks, thereby decreasing the transition temperature. An explanation of why the tungsten-boron alloys did not decrease the tensile transition temperature is not available at this time.

SUMMARY OF RESULTS

The effects of selected alloying elements on the high-temperature creep

and tensile strength and the ductile-brittle transition behavior of arc- and electron-beam-melted tungsten were investigated and gave the following results:

1. The tensile and creep strength of arc- and electron-beam-melted tungsten above 2500° F can be significantly increased by additions of hafnium, tantalum, columbium, and rhenium (in order of decreasing effectiveness). A boron addition of 0.25 atomic percent produced a marked initial strengthening, but larger additions produced only moderate additional strengthening.

2. Columbium, tantalum, and hafnium strengthen both by solid-solution formation and by reacting with residual interstitials to form fine dispersions.

3. The addition of carbon to a nominal tungsten 1-atomic-percent-columbium alloy resulted in greatly increased high-temperature strengthening attributed, in part, to a stabilization of the cold-worked structure of the alloy. A tungsten 1.15-atomic-percent-columbium 1.32-atomic-percent-carbon alloy had a tensile strength of 72,600 psi at 3000° F, which is higher than the value reported for any other metallic material at this temperature.

4. Additions of 2.11 and 6.64 atomic percent rhenium to electron-beam-melted tungsten resulted in 4t bend-transition temperatures below room temperature for 0.40-inch-thick sheet in the stress-relieved condition.

5. Additions of boron and rhenium reduced the bend-transition temperature of recrystallized arc-melted tungsten 250° to 350° F below that of unalloyed tungsten.

6. Rhenium decreased the temperature dependency of the yield stress, permitting the alloys to yield prior to fracture at temperatures lower than that for unalloyed tungsten.

Lewis Research Center
National Aeronautics and Space Administration
Cleveland, Ohio, October 12, 1964

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